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SODIUM CONCENTRATION MEASUREMENTS USING LASER INDUCED FLUORESCENCE

THESIS

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Vincent N. Benham Captain USAF

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SODIUM CONCENTRATION MEASUREMENT USING LASER INDUCED FLUORESCENCE

THESIS

Presented to the Faculty of the School of Engineering
of the Air Force Institute of Technology
Air University
in Partial Fulfillment of the
Requirements for the Degree of
Master of Science

by

Vincent N. Benham

Captain

USAF

Graduate Engineering Physics

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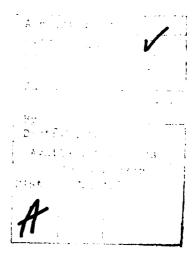
Preface

This thesis effort was a very valuable experience.

The knowledge and experience gained in using information
learned in the classroom is immeasurable. The many hours
of frustration was time well invested.

My sincere appreciation goes to my thesis advisors,
Dr. Robert Hengehold and Dr. Won Roh. Their patience and
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Contents

		Page
Pref	ace	ii
List	of Figures	iv
Abst	ract	v
I.	Introduction	1
II.	Theory	4
	Fluorescence Signal	4 8 11 12
III.	Equipment	14
	LaserOpticsSample HolderElectronics	14 17 18 18
IV.	Procedure	20
v.	Results and Discussion	22
	Uncertainty	22 23
VI.	Conclusions and Recommendations	27
	Conclusions	27 27

List of Figures

Figure		Page
1.	Hyperfine Structure of Sodium D Lines	5
2.	Two Level Model	5
3.	Photon Counting System	11
4.	Three Level Model	13
5.	Experimental Setup	15
6.	Fabry Perot Interference Pattern	16
7.	Vacuum Chamber	18
8.	PHA Mode of Discriminater	19
9.	Table of Data ······	24

Abstract

An experimental system to detect small concentrations of atoms and molecules through the method of laser induced fluorescence is assembled and characterized. A tunable dye laser is used as the excitation source, and sodium is used as a test sample. Sodium is detected in concentrations down to 3.79×10^3 cm⁻³.

SODIUM CONCENTRATION MEASUREMENTS USING LASER INDUCED FLUORESCENCE

I. Introduction

Low concentrations of atomic species can be detected using laser-induced fluorescence. This technique has been used to detect small amounts of atomic sodium (Na) (Ref 1-5). Fluorescence is the emission of light from the decay of an atom or molecule in an excited state. The atom is excited through an inelastic collision with a photon of energy hu which equals the energy of the atomic transition. After a lifetime characteristic of the excited state, the atom will relax to the ground state with the spontaneous emission of a photon. If the emitted fluorescence has the same frequency as the incident light it is called resonance fluorescence. It is very sensitive for detecting low concentrations because many photons per second can be scattered from each atom. But because the emitted photons have the same frequency as the stimulating light it is not possible to distinguish signal and stray-light photons. Therefore, stray light must be kept to a minimum or else the signal must be modulated to make it distinguishable from the background stray-light.

The advent of the tunable dye laser has solved many of the problems associated with atomic and molecular fluorescence analysis (Ref 6:168-172). The dye laser is tunable over the visible region. It is spatially and temporally coherent which can allow high power densities and narrow line widths. As a result, laser induced fluorescence promises high detection

sensitivities plus a high degree of specificity in species detected. A very high degree of spatial resolution of the atomic concentration is also possible due to the fact that only the atoms that are in the laser beam and are viewed by the optics used will be detected.

There are many applications where spatially resolved species concentration measurements are required. One is in the area of microwave devices. The lifetime of high power microwave tubes is limited primarily by the degradation of oxide cathodes used in the tubes. However, the degradation mechanism of the cathodes is not well understood at the present time. It is generally believed that upon activation the oxides of Ba or Ca present in the cathode matrix breakdown and allow a monolayer of metallic Ba to coat the tungsten cathode material. This results in a reduced work function and thus increased thermionic emission. As the cathode ages, the supply of Ba is used up until it is finally depleted, at which time the cathode fails. Where the Ba goes, what becomes of the oxygen, and what effect impurity atoms have on the process is not well understood. An analysis of the atomic species leaving the cathode surface under actual operating conditions should provide insight into the process of activation, emission, and aging (Ref 7).

The purpose of this thesis effort is to develop and optimize a system capable of detecting and measuring small concentrations of atomic and molecular species through laser-induced resonance fluorescence. Because the emitted photons have the same frequency as the laser, background

scatter must be minimized. Since the overall goal is to detect small concentrations, the emitted photons must be distinguished from noise in the photon counting system.

Noise comes from scattered photons, dark current from the photomultiplier tube, random generation of charge carriers, and fluctuations in voltage across circuit elements.

Sodium is chosen to be detected in this effort because it is easy to introduce into the system and easy to detect. It is anticipated that the system will require a minor modification to detect barium atoms emitted from an activated cathode in a subsequent effort.

The scope of this thesis is limited to the development of a system capable of measuring small concentrations of sodium atoms and characterizing this system.

The theory of fluorescence and photon counting are discussed in Chapter II. The equipment used is described in Chapter III followed by a discussion of the procedures in Chapter IV. Chapter V gives a summary of the results, and Chapter VI gives conclusions and recommendations.

II. Theory

The physical theory which forms the basis for the experiment is discussed in this section. The equations used for determining species concentration from a detected fluorescence signal are developed, followed by the development of a method for calibrating the photon-counting system.

Finally a brief description of photon-counting is presented.

Fluorescence Signal

The sodium D lines are composed of transitions between hyperfine levels as seen in Fig. 1. The D₁ line corresponds to $P_{\frac{1}{2}}$ -S_{$\frac{1}{2}$} transition and the D₂ line corresponds to the $P_{\frac{1}{2}}$ -S_{$\frac{1}{2}$} transition. The spectral profile of these transitions can be determined by summing up the contributions of the individual Doppler broadened hyperfine transitions. Using the relative transition strengths shown in Fig. 1, one can show that the absorption curves have nearly identical shape but the D₂ line has twice the magnitude of the D₁ line. For this reason the dye laser was tuned to the D₂ line in sodium.

The sodium atoms are pumped to the $P_{\frac{3}{2}}$ energy level by the laser. They are maintained in an ultrahigh vacuum so that collisional excitation can be ignored. The $P_{\frac{3}{2}}$ - $P_{\frac{1}{2}}$ transition is forbidden. The excited atoms decay to the $S_{\frac{1}{2}}$ state. Therefore the D_2 transition can be modeled by the two level system shown in Fig. 2. The energy separation of the two levels corresponds to that of the $P_{\frac{3}{2}}$ - $S_{\frac{1}{2}}$ transition.

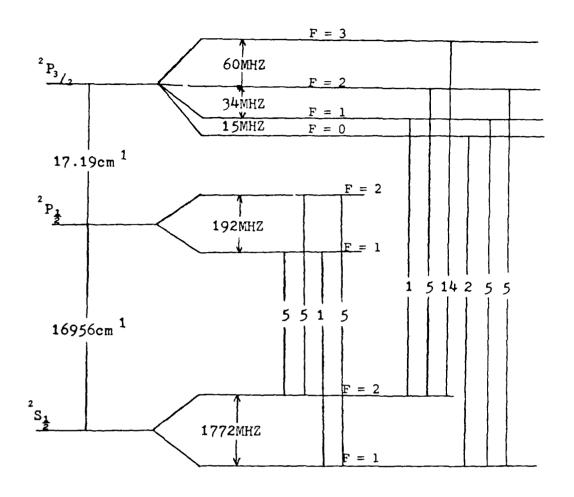


Fig. 1. Hyperfine Structure and Transition Strengths of the Sodium D Lines. (Ref 2:201)

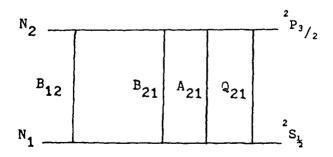


Fig. 2. Two-Level Model

If the sodium atoms are irradiated by a laser of spectral energy density $\rho(\text{joules/m}^3-\text{hz})$ and if ρ is constant over the spectral profile of the atomic transition then the transition rate equation may be written

$$\frac{dn_2}{dt} = -\frac{dn_1}{dt} = N_1 B_{12} \rho - N_2 (B_{21} \rho + A_{21} + Q_{21})$$
 (1)

where $\rm N_2$ and $\rm N_1$ are the upper and lower state number densities, $\rm B_{12}$ and $\rm B_{21}$ are the Einstein coefficients for stimulated absorption and emission, $\rm A_{21}$ is the Einstein coefficient for spontaneous emission, and $\rm Q_{21}$ is the quenching rate of the atoms in the upper state due to collisions with other atoms or molecules.

In addition

$$N_1 + N_2 = N_T \tag{2}$$

where $\mathbf{N}_{\mathbf{T}}$ is the total number density of atoms in the sample volume.

If ρ is constant in time then the steady state condition $dN_2/dt = 0 \text{ applies so that combining Eq (1) and Eq (2)}$

$$\frac{N_2}{N_T} = \frac{B_{12} \rho}{(B_{12} + B_{21}) \rho + A_{21} + Q_{21}}$$
 (3)

For large spectral energy density such that $(B_{12}+B_{21})$ $\rho >> A_{21}+Q_{21}$ the transition is called saturated and

$$\frac{N_{2}}{N_{T}} = \frac{B_{12}}{B_{12} + B_{21}} = \frac{1}{1 + \underline{g}_{1}}$$
(4)

where g_1 and g_2 are the degeneracies associated with the lower and upper states respectively and where

$$B_{12}g_1 = B_{21}g_2 \tag{5}$$

has been used.

Eq (3) can be rewritten as

$$\frac{N_2}{N_T} = \frac{B_{12}\rho}{(B_{12}+B_{21})} \rho \cdot \frac{1}{\frac{A_{21}+Q_{21}}{(B_{12}+B_{21})} \rho^{+1}}$$

$$= \frac{g_2}{g_1+g_2} \cdot \frac{\left(\frac{B_{12}+B_{21}}{A_{21}+Q_{21}}\right) \rho}{1+\left(\frac{B_{12}+B_{21}}{A_{21}+Q_{21}}\right)} \rho$$
(6)

$$= \frac{N_2}{N_T}$$
 sat $\cdot \frac{R}{R+1}$

where the saturation parameter R is defined as

$$R = \left(\frac{B_{12} + B_{21}}{A_{21} + Q_{21}}\right) \quad \rho \tag{7}$$

and its value is an indication of the degree of saturation. For large values of R, N_2/N_T approaches its saturation value. The value of R can be found from the value of the spectral energy density if the Einstein coefficients are known. (The quenching term Ω_{21} can be ignored in this effort because of the high vacuum.) It can also be measured experimentally. Under extreme saturation conditions the ratio of atoms in the upper state to those in the lower state approaches the ratio of the degeneracies of the two states.(Ref 8). Since R is proportional to the laser power and fluorescence is proportional to N_2 , the difference in fluorescence signal can be noted with a known change in laser power and R can be determined using Eq (6).

The fluorescence signal detected by the photon counting system arrives as a result of the spontaneously emitted photons when the atoms make a transition from the upper state to the lower state. Assuming that the photons are emitted isotropically, the detected fluorescence signal is given by the following equation.

$$I_f = hv A_{21} N_2 \frac{\Omega}{4\pi} VK$$
 (8)

where ho is the energy separation of the two energy levels, Ω is the solid angle defined by the detection optics, and V is the volume of fluorescing atoms seen by the detection system. K is the total efficiency of the detection system. It includes all the losses due to windows, lenses, filters, misalignment, plus the guantum efficiency of the photomultiplier tube (PMT). Eq (8) can be rewritten using equations (4) and (6) as

$$I_{f} = hvA_{21} \frac{N_{T}}{1+g_{1}} \frac{\Omega}{4\pi} VK \frac{R}{R+1}$$
 (9)

In principle, N_T , the total concentration can be gotten using Eq (9) since all other terms are known or experimentally determined. However, misalignment, unknown losses and uncertainty in quantum efficiency can affect the values of Ω , V, and K. These values can be determined experimentally using the Rayleigh scattering of a known concentration of gas. A method is derived on the following pages to experimentally determine Ω , V, and K and therefore calibrate the experimental system.

Calibration

If a molecule is placed in a sinusoidally alternating electric field it radiates as a dipole with an intensity given as (Ref 9:101)

$$\frac{\mathrm{d}\Phi}{\mathrm{d}\Omega} = \frac{\omega^4 \sin^2\Theta}{32\pi^2 \varepsilon_0 c^3} \sum_{\epsilon_0}^{-2} E_0^2$$
 (10)

If there are N_{R} molecules radiating incoherently the expression for the average radiated intensity becomes

$$\frac{d\Phi}{d\Omega} = \frac{\omega^4}{32\pi^2 \epsilon_0 C^3} N_R \overline{\alpha}^2 E_0^2$$
 (11)

where ω is the angular frequency of the electric field, E_0 is its amplitude, and $\overline{\alpha}$ is the average polarizability, ε_0 is the permittivity for free space, and c is the speed of light. If the electric field is due to a laser then Eq (11) can be written as

$$\frac{d\Phi}{d\Omega} = \frac{\pi^2}{\varepsilon_0^2 \lambda^4} \,^{N}_{R} \,^{\alpha^2} \,^{\ell}$$
 (12)

where

$$\ell = \frac{1}{2} c \epsilon_0 E_0^2 \tag{13}$$

is the irradiance of the incident beam and λ is the wavelength. Using the definition of a scattering cross section

$$\frac{d\Phi}{d\Omega} = N_R \frac{d\sigma}{d\Omega} \ \varrho \tag{14}$$

and comparing this with Eq (12) leads to the conclusion that

$$\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega} = \frac{\pi^2}{\epsilon \alpha^2 \lambda^4} \overline{\alpha}^2 \tag{15}$$

And since the measured Rayleigh signal from a gas is

$$I_{R} = N_{R} \frac{d\sigma}{d\Omega} \ell V\Omega K$$
 (16)

then

$$V\Omega K = \frac{I_R}{N \frac{d\sigma}{d\Omega}} \chi \tag{17}$$

Since $V\Omega K$ can remain the same for a flourescence measurement and a Rayleigh scattering measurement with another gas, its value can be determined from Eq (17) and (15) if $\overline{\alpha}$ can be

derived. The polarizability can be related to the index of refraction as shown below:

From electromagnetics

$$D = \varepsilon_0 kE$$

$$= \varepsilon_0 E + P$$

$$= \varepsilon_0 E + N = E$$
(18)

where D is the dielectric displacement and P is the polarization. Therefore

$$k = 1 + \frac{P}{\varepsilon_0}E$$

$$= 1 + \frac{N\overline{\alpha}}{\varepsilon_0}$$
(19)

And

$$k = n^2 \tag{20}$$

where n is the index of refraction of the medium. Therefore from Eqs (17) and (19)

$$\frac{1}{\alpha} = \frac{\epsilon_0}{N} \quad (n^2 - 1) \tag{21}$$

Now rewriting Eq (9)

$$N_{T} = \frac{4\pi \left(1 + \frac{\alpha_{1}}{g_{2}}\right)}{h\nu A_{21}} \qquad \boxed{\frac{1}{V\Omega K}} \qquad \boxed{\frac{R+1}{R}} I_{f} \quad (22)$$

which gives the total number density as a function of the detected fluorescence signal. We can substitute the experimental value $V\Omega K$ determined from the Rayleigh scattering measurement using Eqs (17), (15), and (21)

$$N_{T} = \frac{4\pi \left(1 + \frac{g_{1}}{g_{2}}\right)}{h\nu A_{21}} \begin{bmatrix} N_{R} \ell \pi^{2} \left(\frac{\epsilon_{0}}{N_{R}} \left[n^{2} - 1\right]\right)^{2} \\ \frac{1}{R} \epsilon_{0}^{2} \lambda^{4} \end{bmatrix} \begin{bmatrix} R+1 \\ R \end{bmatrix} I_{f}$$
 (23)

From Eq (23) the concentration of sodium atoms can be determined using experimental data and some well known constants.

Photon-Counting

The most sensitive technique for measuring small fluorescence signals is to count the photons striking the detection system. A schematic of a photon-counting system is shown in Fig. 3.

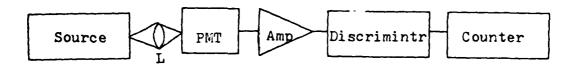


Fig. 3. Photon-Counting System

Some of the photons striking the photo-cathode of a photo-multiplier tube results in voltage pulses across its anode load resistor. These pulses are amplified and sent to a discriminator. Only those amplified pulses whose amplitudes are larger than the discrimination level that has been set on the discriminator generate square pulses which are then counted.

The ratio of striking photons to electrons emitted from the photo-cathode is called the quantum efficiency. The emitted photons travel down a chain of dynodes of increasingly positive potentials. The energy acquired in traveling to the next dynode causes several secondary electrons to be emitted by the struck dynode and hence there is a gain at each dynode. It is desirable that the dynode gain distributions

be Poissonian and that the first dynode gain be as large as possible. This will help keep the total gain of the photomultiplier tube constant. (Ref 10:1-9)

Photo-electrons emitted from the photo-cathode of the PMT cannot be distinguished from thermionic electrons emitted by the photo-cathode. Thermionic emissions are present even in the dark but can be minimized by cooling the PMT. The dynodes also emit thermionic electrons but because they do not experience as much gain their resulting output pulses can be stopped at the discriminator along with dc leakage currents and other electronic noise.

If the voltage applied to the PMT is increased, the count of photo-emitted electrons will increase and then level out and tend not to increase much further with highter voltage. However, the dark count may continue to rise with the voltage. Thus there is an optimum bias voltage for the highest signal to noise ratio.

Three level System

The equations derived earlier in this chaper pertain to a two level system like the D lines of sodium. Other atoms of interest may be modeled as a three level system shown in Fig. 4.

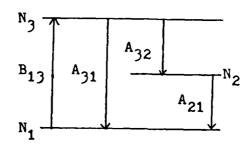


Fig. 4. Three-Level Model

The laser would be tuned to the wavelength corresponding to the transition from level 1 to level 3. This would excite the atoms into level 3. From there they would spontaneously decay to the second and first levels.

In a similar manner as done earlier in this chapter, Daily (Ref 11:569) has shown that for a three level system

$$N_{T} = \begin{pmatrix} 1 + \underline{q}_{1} \\ \overline{q}_{3} \end{pmatrix} \frac{\underline{I}_{3}}{h_{V}(A_{31}/4\pi) V\Omega K} + \frac{\underline{I}_{2}}{h_{V}(A_{21}/4\pi) \Omega V K}$$
(24)

where I_3 is the fluorescence signal resulting from a transition from level 3 to level 1 and I_2 is the fluorescence signal resulting from a transition from level 2 to level 1. I_3 and I_2 have different wavelengths and must be distinguished from each other possibly through the use of a monochrometer.

As before, the Rayleigh scattering calibration technique may be used to derive $V\Omega K$.

III. Equipment

The equipment used for this effort is divided into the following categories: (1) laser, (2) optics, (3) sample holder, and (4) electronics. Each of these categories will be discussed separately. A schematic of the overall experimental setup is shown in Fig. 5.

Laser

A Spectra-Physics Model 164 Argon Ion Laser with a maximum output of 1.5 watts emitting at 514.5nm is used to pump a Spectra-Physics Model 370 Tunable Dye Laser. The dye laser is modified with a solid etalon of 1mm thickness positioned in the center of the cavity. This narrowed the line width from 30 GHz without the etalon to 13 GHz with the etalon in position. Course tuning of the emitted wavelength is accomplished by rotating the intracavity dispersing prism. Fine tuning is accomplished by tilting the intracavity etalon.

The dye laser can be tuned to resonance with the sodium D_1 or D_2 line with the aid of the sodium lamp shown in Fig. 5. A portion of the beam is split off and passed through the lamp which is turned on. As the laser is tuned through the D_1 and D_2 lines, the sodium atoms can be seen fluorescing in the lamp.

The spectrum of the laser is analyzed with a fixed Fabry
Perot etalon. The resulting interference pattern shows
evidence of another etalon in the cavity that is approximately
2.5cm in thickness. (Fig 6) It appears that the dye laser

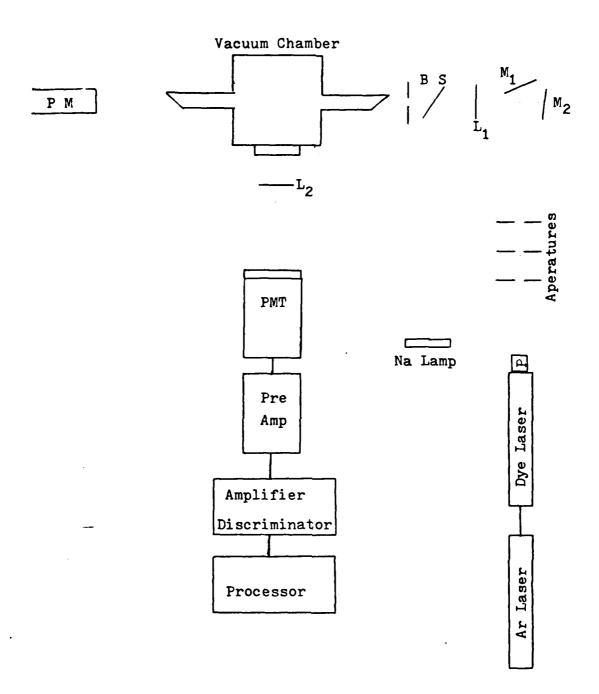


Fig. 5. Experimental Setup

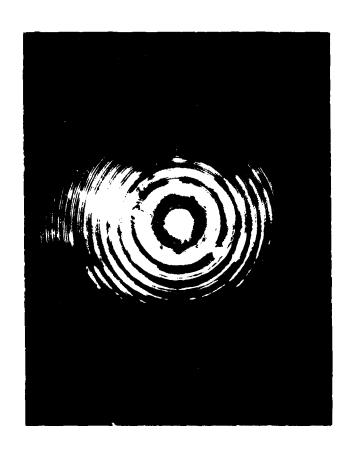


Fig. 6. Fabry Perot Interference Pattern

input mirror and a window of the dye cell form this etalon. These two components are approximately 2.5cm apart and the tilting of the dye cell can affect the frequency output of the laser enough to take it in and out of resonance with the sodium D line.

A Spectra Physics Model 401C Power Meter is used to observe laser power.

Optics

The beam leaves the laser polarized in the vertical direction. It passes through a polarizer to insure this polarization. A series of apertures clean up the profile of the beam. Two mirrors are used to turn the beam in order to guarantee that the beam at M_2 can be placed in line with the center line of the vacuum chamber. Tuning the laser affects the beam position to a small degree, and as will be shown in the next section, the beam position was critical. The beam then travels through lens L_1 to focus it to the center of the vacuum chamber, it has a focal length of 50.8cm. The vacuum chamber is fitted with brewster windows to reduce scattered light.

Photons emitted from flourescing sodium atoms in the vacuum chamber can pass through a window and the second lens L_2 (10cm in diameter) that is two focal lengths (23cm) away from the center of the laser beam. The beam is then imaged on a slit in front of the PMT that is two focal lengths on the other side. A 5890 Å filter with a 3 Å FWHM bandpass is positioned in front of the PMT to reduce unwanted light.

Sample Holder

A cutaway view of the vacuum chamber is shown in Fig. 7.

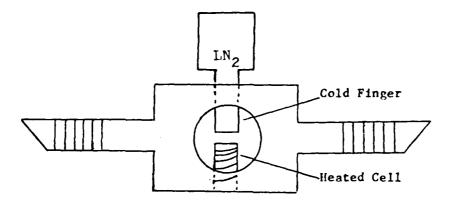


Fig. 7. Vacuum Chamber

A series of 6 annodized aluminum baffles with ½ inch diameter concentric holes are aligned in each arm of the vacuum chamber. The baffles reduce scattered light. Metalic sodium is placed in a cell (Fig. 7) which is heated by heater wires encircling it. Directly above the heated cell is a cold finger cooled with liquid nitrogen to condense the sodium vapor. The back chamber wall is covered by an annodized aluminum screen to reduce the chance of scattered light being reflected towards the PMT.

Pressure was maintained at $2x10^{-8}$ torr with the cold finger and $1x10^{-7}$ torr without it.

Electronics

The electronics is composed of the PMT, amplifier-

discriminator and counter. The amplifier discriminator is a Princeton Applied Research Model 1121, the counter is a Princeton Applied Research Model 1112. The PMT is an RCA 8850, which is placed inside a cooler and chilled to 5°C. Tests with a constant optical signal indicated that the best signal to noise ratio could be obtained with a bias voltage between 1900 and 2000 volts. The discriminator threshold level was obtained by placing the discriminator in the pulse height analyzer mode and positioning the threshold marker in the valley between the noise and signal as represented in Fig. 8.

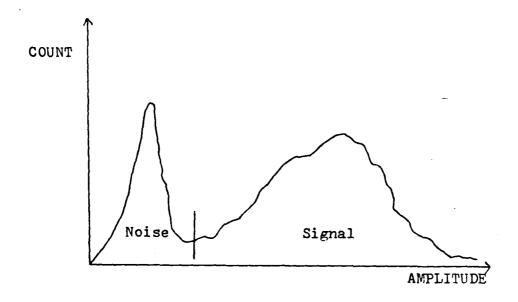


Fig. 8. PHA Mode of Discriminator

IV. Procedure

The simple procedure for making fluorescence intensity measurements is described below.

The vacuum chamber is pumped down to the low 10^{-7} torr range. It is then backfilled with one atmosphere of gaseous nitrogen (N_2) . The N_2 is used for two purposes. It is used to produce the Rayleigh scattering for measuring $V\Omega K$ as derived in Chapter II. At the same time it is used as a light source to align the optics between the vacuum chamber and the PMT.

The region between the vacuum chamber and the PMT is shielded optically such that there is no increase in counts registered from the PMT over the dark count when the laser is off. The laser is turned on and tuned to the sodium \mathbf{D}_2 line with the aid of the sodium lamp and its power noted. The Rayleigh scattered light is used to align the optics for maximum signal. The counts coming from the PMT now are composed of dark count, scattered light, and Rayleigh signal.

The vacuum chamber is then pumped down to the low 10^{-8} torr range. Counts coming from the PMT are now composed of dark count plus scattered light (Background). The line width of the laser is determined with a Fabry Perot etalon. Current is applied to the heater to produce sodium vapor which leaves the cell and condenses on the cold finger after passing through the laser beam. The increase in counts over the background is recorded as fluorescence signal.

The laser can be tuned in and out of resonance with the D_2 line. The counts measured while the laser is out of resonance equal the dark count plus scattered light measured previously.

V. Results and Discussion

The results of five experimental runs are shown in Table I. The slit size leading to the PMT was varied along with laser power. These two parameters have an impact on the background counts. For the last three columns, the voltage applied to the focusing grid between the photo cathode and first dynode was adjusted for maximum efficiency. R_{calc} was derived from Equation 7 after the spectral energy density is determined experimentally. Rexp was determined experimentally using Equation 6 by noting the change in fluorescence signal with change in laser power. VNK was determined experimentally with the method described in Chapter II using Equation 14. $N_{\tau\tau}$ is derived using Equation 23. It is the number density of sodium atoms that can be detected when the ratio of signal to noise equals one. N_{atoms} is the number of atoms in the volume viewed by the detection system. It is the number density times the laser beam volume observed by the PMT. This volume is defined by the laser beam size and slit dimensions.

Uncertainty

As in Chapter IV, background counts are defined as those caused by scattered photons in the vacuum chamber plus dark count. The average background count is listed in Table I for each experimental run. The fluctuation in this background is the noise that limits the concentration of sodium atoms that can be detected. For this study the noise is defined as two times the standard deviation (2g) in the

background count. In other words when the counts on the counter reach the average background plus 20 the sodium atoms are said to be detected in the concentration given in Table I. Obviously, the undertainty in this measurement is ± 100% but it is the absolute upper limit in the system's sensitivity. As the sodium concentration grows beyond this limit the noise becomes less and less significant. At higher concentrations other terms contribute to uncertainty in concentration measurements. The Rayleigh scattered signal used to calibrate the photon-counting system is subject to the same noise in background count. The irradiance of the beam used to make the Rayleigh scattered measurement is known within 8%. The concentration of N2 molecules is assumed to be 1 atmosphere and is therefore known to about 5%. The uncertainty at any concentration is given by the following expression:

$$\Delta N_{T} = N_{T} \left[\left(\frac{\Delta I_{f}}{I_{f}} \right)^{2} + \left(\frac{\Delta Q}{Q} \right)^{2} + \left(\frac{\Delta N_{R}}{N_{R}} \right)^{2} + \left(\frac{\Delta I_{R}}{I_{R}} \right)^{\frac{1}{2}} \right]^{\frac{1}{2}}$$
(24)

Assumptions

The detected fluorescence signal is given by the following equation:

$$I_f = h_V A_{21} N_2 \frac{\Omega}{4\pi} Vk \frac{R}{R+1}$$
 (25)

This was used in Chapter II to derive Equation 23. This assumes that the atoms are radiating istropically in 4π steradians. It also assumes that the laser power is constant over the spectral cross-section of the atomic transition. But as was noted in Chapter III, the laser spectral structure

Slit Size (cm)	.2x.6	.07x.26	.07x.26	.07X.26	.06X.14
Laser Power (mW)	12	18	89	18	18
Background	1.52x10 ³	226	1X10 ³	780	520
Rcalc	5.82	7.80	8.01	8.01	8.01
в вхр	3.98	6.21	5.98	6.01	6.03
VΩK	1.43X10-15	3.2X10-16	3.05x10 ⁻¹⁵	5.31X10-15	2.63X10 ⁻¹⁵
$N_{\rm T}(S/N=1)/cm$	2.08x10 ⁴	2.58X10 ⁴	6.68x10 ³	3.73X10 ³	6.26x10 ³
Natoms	3,55	1.92	. 495	.280	. 248

was not constant but had two distinct peaks. Therefore the calculated value of R is inappropriate since it assumes constant power over the spectral width of the laser line. That is why the experimental value of R was used to calculate $N_{_{\rm T}}$ in Table I.

As shown in Table I, the smallest concentration detectable is 3.73×10^3 while the smallest number of atoms detected is .248. These two numbers did not occur for the same slit size. The concentration detected is a function of the noise and collection efficiency (V Ω K) while the number of atoms detected is only a function of the volume once the concentration has been determined.

After the previous data had been taken it was found that the fluorescence signal was neither randomly polarized nor completely linerly polarized. The degree of polarization (P) is defined as

$$P = \frac{I_1}{I_{11}}$$

where $I_{\underline{1}}$ is the fluorescence signal polarized perpendicular to the polarization of the laser and I_{11} is the fluorescence signal polarized parallel to the polarization of the laser. This value was measured to be .479.

The polarized signal indicates that there is some anisotropic resonance scattering and the determined number densities ($N_{\rm T}$) in Table I should be multiplied by the following factor. (Ref 9:47-49)

$$\frac{2}{3} \left[\frac{1 + 2P}{1 + P - (1 - P)\cos^2 \theta} \right] = .883$$

This factor is the ratio between the fluorescence signal originating from an isotropic scatterer and one originating from an anisotropic scatter. Θ is measured from the polarization of the laser and is 90° for this study.

VI. Conclusions and Recommendations

Conclusions

The following conclusions summarize this experimental effort:

- 1. A system was assembled that can be used to detect low concentrations of atoms and molecules.
- 2. The experimental set-up as designed and built can be used to detect sodium atoms with a density as low as 3.73×10^3 cm⁻³.
- The experimental set-up can detect an average of
 248 sodium atoms in the volume observed.

Recommendations

Based upon the experience of this effort, the following recommendations are made:

- 1. The dye laser should be modified to eliminate the dye cell window and replace it with a dye jet. This would eliminate the etalon effect caused by the cell window.
- 2. A portion of the beam should be split off, its power converted to frequency and fed into the counter/ processor. This could be used to determine the consistency of laser power in addition to making signal to power measurements.
- 3. The system should be modified to be able to heat an impregnated cathode and look for the emitted barium or barium oxide.

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VITA

Vincent N. Benham was born on 6 December 1953. He attended Wichita State University in Wichita, Kansas after graduating from high school in 1972. In 1976 he received the degree of Bachelor of Arts in Physics and a commission in the United States Air Force. He was assigned to the Rocket Propulsion Laboratory at Edwards Air Force Base where he worked on electric propulsion for satellites.

Captain Benham began his tour at the Air Force Institute of Technology in June 1980. He is married to the former Kim Christensen and has two daughters, Jennifer and Sara.

Permanent address: 123 N. Sedgwick Wichita, Kansas 67203

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An experimental system to detect small concentrations of atoms and molecules through the method of laser induced fluorescence is assembled and characterized. A tunable dye laser is used as the excitation source, and sodium is used as a_3 test sample. Sodium is detected in concentrations down to 3.79×10^3 cm⁻³.

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